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Metamaterials: *supra*-classical dynamic homogenization

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PAPER

Metamaterials: *supra*-classical dynamic homogenization*

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E-mail: Mihai.Caleap@bristol.ac.uk**Keywords:** metamaterials, dynamic homogenization, self-consistent effective field methodSupplementary material for this article is available [online](#)

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Abstract

Metamaterials are artificial composite structures designed for controlling waves or fields, and exhibit interaction phenomena that are unexpected on the basis of their chemical constituents. These phenomena are encoded in effective material parameters that can be electronic, magnetic, acoustic, or elastic, and must adequately represent the wave interaction behavior in the composite within desired frequency ranges. In some cases—for example, the low frequency regime—there exist various efficient ways by which effective material parameters for wave propagation in metamaterials may be found. However, the general problem of predicting frequency-dependent dynamic effective constants has remained unsolved. Here, we obtain novel mathematical expressions for the effective parameters of two-dimensional metamaterial systems valid at higher frequencies and wavelengths than previously possible. By way of an example, random configurations of cylindrical scatterers are considered, in various physical contexts: sound waves in a compressible fluid, anti-plane elastic waves, and electromagnetic waves. Our results point towards a paradigm shift in our understanding of these effective properties, and metamaterial designs with functionalities beyond the low-frequency regime are now open for innovation.

Introduction

Metamaterial research in the past decade offered an entirely new route to further enhance our capability to engineer material properties at will. Here, metamaterials are artificially fabricated structures (often periodic, i.e., crystalline) which are designed so that they exhibit wave properties not observed with common materials, e.g., they can, in theory, bend electromagnetic [1], acoustic [2], and even surface gravity waves [3] so as to achieve sub-wavelength focusing [4], create cloaks [5, 6], and attain shielding [7]. Other unexpected properties include artificial magnetism [8], negative permeability [9], negative refraction index [10], and hyperbolic dispersion [11], to name a few. Such materials have allowed us to gain unprecedented control over a range of electromagnetic/optical and acoustic wave phenomena. In many ways metamaterials parallel the development of photonic and phononic crystals (optical and acoustic analogues of semiconductors) which also rely on small-scale structures for their properties. However, the major difference lies in the sub-wavelength nature of metamaterial structure. This enables us to summarize their properties in terms of permittivity and permeability (ϵ , μ) for electromagnetic waves, or bulk modulus and mass density (κ , ρ) for acoustic waves, just as we would for any naturally occurring material. This is an enormous simplification for the design process, and research is now focusing on the realization of a new generation of metadevices [12] with novel and useful functionalities achieved by the structuring of functional matter on the sub-wavelength scale. Novel devices such as superlens [13], hyperlens [14], invisibility cloaks [15, 16], and plasmonic waveguides [17] have been fabricated and tested in the past few years. The technology behind such metadevices is fairly well established in the *low-frequency* regime where inclusions have sizes much smaller than the wavelength of operation. At these relatively low frequencies this is commonly obtained by assuming only monopole and/or dipole interactions, e.g., by utilizing

* Dedicated with gratitude to the memory of Prof Yves C Angel.

conducting materials shaped as dipoles [18] and split-ring resonators [19]. The existence of resonances poses a considerable challenge to classical effective medium theories. This is because their basic principle is to minimize the scattering in the quasi-static limit, while the local resonances occur most often at longer wavelengths.

Here, following these concepts, we develop and analyse a *supra*¹-classical dynamic model of metamaterial response. There is an abundance of miscellaneous effective medium theories [20–26], some quite recent [27–29]; many of these works claim to be valid not only in the quasi-static limit but also at finite frequencies *beyond* the long-wavelength limit: a situation that happens when the wavelength Λ is long in the host medium, while the wavelength in the particles, Λ_0 , can be small. (This is in contrast to the quasi-static limit where both Λ and Λ_0 should be much larger than the size of the particles.) Such extension to finite frequencies is sometimes denoted as the dynamic effective medium theory. However, even this *dynamic* approximation relies exclusively on the monopolar and dipolar response of the scattering objects, which implicitly assumes long wavelengths. In this paper, this restriction is relaxed and the full effect of the ensemble of particles that constitute the effective medium is included, as higher diffraction orders are encompassed. This will allow the design of new metadevices working over a wider wavelength range. We shall illustrate this by solving a simple scalar problem in two-dimensions, having applications not only in electromagnetics but also in acoustics and elasticity. The similarities between the equations of acoustics, elasticity and electromagnetics allow us to use some of the same techniques to solve problems in these seemingly disparate fields.

View on classical homogenization

The theoretical approach to the field of metamaterials is provided through dynamic homogenization techniques which relate the microstructure of a composite to the frequency dependence of its effective properties. The majority of research interest in the area of metamaterials is restricted to *periodic* microstructures [30, 31] (as the arrangement of molecules according to solid-state physics) which admit Bloch (or Floquet) waves as solutions and many different numerical algorithms have been developed (see, e.g., [32, 33]) for calculating the dispersive properties of these waves. A popular route to determining these parameters is by the use of retrieval methods [34, 35] where the assumption is that local effective properties may be used to define periodic composites. The retrieval method leads to the refractive index n and the wave impedance \mathcal{Z} , which defines the reflectivity of a semi-infinite slab. However, while simple in principle, such retrieval methods are limited to ordered arrays and often produce ambiguous results due to oversimplified initial assumptions of the bulk model [36].

Certainly engineers like structures and designs that follow some type of order. However, materials may be also amorphous and isotropic, and natural materials on the macroscopic level are quite often *random* in essence. It may well be that a random placement of complex particles would be enough to produce emergent properties in the overall wave response and therefore give us a sample of metamaterial [37]. The effective behavior of metamaterials whose microstructure is random depends strongly on the governing statistics of the random distribution. Effective properties may be determined by using the self-consistent *effective medium* methods for which a substantial body of literature may be found. Although variants exist, these methods often consider the scattering problem of a coated particle embedded in a matrix which has the properties of the effective media. These properties are then determined by requiring the vanishing of the effective forward-scattering amplitude $f_0^{\text{eff}} = f_{\theta=0}^{\text{eff}}$ and as such are formally restricted to the low-frequency and long-wavelength ranges. For examples where this method has been applied to electromagnetic, acoustic and elastic waves, see [20–22, 28, 38, 39]. Although the above self-consistent condition ($f_0^{\text{eff}} = 0$) is physically sufficient to describe the effective medium, two effective properties, i.e. $(\epsilon_{\text{eff}}, \mu_{\text{eff}})$, cannot be determined ‘simultaneously and uniquely’ from the single condition. A supplementary condition is needed; this prevents the application of effective medium methods to finding dynamic effective properties. Note however that the above condition is sufficient for wave propagation in a metamaterial in which a single material constant is involved, e.g. in dielectric media. Another deficiency of many current enhancements of the effective medium methods is their failure to describe the influence of the spatial distribution of particles on the effective constitutive parameters. Such a description is possible in the framework of a self-consistent scheme called the *effective field* method [37] and our work is within the framework of this scheme. One of the principal results of the effective field approach was an adequate definition of the coherent wave and a proof that it obeys a wave equation, i.e., a proof that, under certain conditions, a random distribution of scatterers can, for this purpose, be represented by an effective medium [40]. Most calculations proceed by assuming the existence of such an effective medium equation.

The subject of the present work is the macroscopic dynamic behavior of the above composite medium, i.e., random distribution of particles. More precisely, we shall describe a heuristic scheme for evaluating the effective properties of metamaterials. The approach is based on the idea that a certain effective field acts on each particle,

¹ This word comes from Latin and means *above* or *beyond the limits of*.

Table 1. Relationships among electromagnetic, acoustic and elastic material parameters.

	Electromagnetics ^a		Acoustics	Elasticity
m	ϵ^{TM}	μ^{TE}	ρ^{P}	$1/G$
d	μ^{TM}	ϵ^{TE}	$1/\kappa$	ρ^{SH}

^a Observe that the permittivity and permeability for a specific polarization can be related to a pair of acoustic and elastic constants. For instance, $(\epsilon^{\text{TE}}, \mu^{\text{TE}}) \leftrightarrow (\rho^{\text{SH}}, \rho^{\text{P}}) \leftrightarrow (1/\kappa, 1/G)$.

as a consequence of the presence of the other particles; hence, the name *effective field* method. The framework we develop is based on the Fikioris–Waterman [41, 42] and Waterman–Pedersen [43] formalism to evaluate the coherent wave motion on both sides of a semi-infinite array of particles². More specifically, we consider an averaged wave motion, where all possible configurations of particles are weighted by appropriate pair-correlation functions. In contrast to the effective medium methods, we derive a fully dynamic model for the effective constitutive parameters, which retains all the relevant information (particle geometry and physical parameters) provided by an expanded multipole solution. As a result, the theory discussed in the following is more complete and potentially more useful than previous approaches to derive effective material parameters.

Results

Here, we consider two specific polarizations in electromagnetism, transverse electric (TE) and transverse magnetic (TM). In addition, parallel to the electromagnetic example is the mathematically identical case of acoustics and anti-plane elasticity.

We then consider these as two-dimensional problems. Indeed, exploiting the physics common to many types of wave propagation, the idea of metamaterials has been implemented successfully for acoustic and elastic waves. Many of the conclusions drawn from photonics research directly apply to acoustic waves and acoustic metamaterials due to the essential similarity of the governing equations in the two cases. Realizing analogous results for elastic metamaterials is complicated by the fact that the governing equation for elasticity admits both longitudinal and shear wave solutions which are capable of exchanging energy between each other. However, anti-plane elasticity is a special state of deformation with just a single non-zero displacement field, similar to transverse electromagnetics. The governing equation common to electromagnetics, acoustics, and linear anti-plane elasticity is detailed in appendix A.

Effective constitutive parameters depend on many factors including the intrinsic properties of the particles and the host matrix, their shape and topology. The latter determines how the particles are distributed in the matrix. The system considered in our study is composed of two isotropic phases: cylindrical particles of arbitrary shape randomly distributed in a host medium with propagation constant $k = \omega\sqrt{m d}$ for some (possibly complex) parameters (m , d) of the medium. Depending on the application, these material parameters could be, e.g., compliance ($1/G$) and density (ρ) for shear horizontal polarized elastic waves or permittivity (ϵ) and permeability (μ) in electromagnetism; a number of useful relationships among these parameters are summarized in table 1.

In appendix B, we briefly review the effective field method. Subject to the quasi-crystalline approximation, two equations are obtained for which the effective wavenumber \mathcal{K} of some coherent wave motion (either electromagnetic, acoustic, or elastic) and the effective impedance \mathcal{Z} , are given in implicit form. Note that, whereas the dispersion relation for \mathcal{K} is polarization-independent, the effective impedance depends on the type of the incident wave. These equations are the starting point of all further developments. Observe that the particles have a size distribution and their relative positions are described by an arbitrary *cross-pair* distribution function g_{ij} . Also, the size distribution is represented by $\eta_j = \eta(a_j)$; here a_j is the radius of the circular surface circumscribing a particle, and η is the number of particles per unit area.

Without loss of generality, we next assume the particles are identical and have equal sizes $a_j \equiv a$. Here, we refer only to the final explicit solutions for the effective parameters m_{eff} and d_{eff} , which are expressed elegantly as

$$\frac{m_{\text{eff}}}{m} \simeq 1 + \tilde{m}_1 \frac{\epsilon}{2k^2} + \tilde{m}_2 \frac{\epsilon^2}{2k^2} + \mathcal{O}(\epsilon^3), \quad (1)$$

² The later reference, is the earliest work to our knowledge to predict explicit relations for the effective bulk parameters (ϵ_{eff} , μ_{eff}) in the dynamic range. The authors have also predicted negative frequency-dependent μ_{eff} at single-particle resonances although the plots only displayed the positive values. In fact, they only noted that ‘the effective parameters vanish or diverge at certain frequencies’ without further comment, which suggests that the results were considered curious at that time. Currently it is common to have negative effective parameters, and much research on metamaterials is focused on this area.

$$\frac{d_{\text{eff}}}{d} \simeq 1 + \tilde{d}_1 \frac{\epsilon}{2k^2} + \tilde{d}_2 \frac{\epsilon^2}{2k^2} + \mathcal{O}(\epsilon^3), \quad (2)$$

where $\epsilon = 4\pi\eta$. The scalar coefficients $(\tilde{m}_1, \tilde{m}_2)$ and $(\tilde{d}_1, \tilde{d}_2)$ are given in matrix notation by

$$\tilde{m}_1 = \mathbf{e}^t \mathbf{Q} \mathbf{e} - \mathbf{e}^t \mathbf{J} \mathbf{Q} \mathbf{e}, \quad (3a)$$

$$\tilde{m}_2 = \mathbf{e}^t \mathbf{Q} \tilde{\mathbf{R}} \mathbf{Q} \mathbf{e} - \mathbf{e}^t \mathbf{J} \mathbf{Q} \tilde{\mathbf{R}} \mathbf{Q} \mathbf{e} - \frac{1}{4k^2} \left[(\mathbf{e}^t \mathbf{Q} \mathbf{e})^2 - (\mathbf{e}^t \mathbf{J} \mathbf{Q} \mathbf{e})^2 \right] \quad (3b)$$

and

$$\tilde{d}_1 = \mathbf{e}^t \mathbf{Q} \mathbf{e} + \mathbf{e}^t \mathbf{J} \mathbf{Q} \mathbf{e}, \quad (4a)$$

$$\tilde{d}_2 = \mathbf{e}^t \mathbf{Q} \tilde{\mathbf{R}} \mathbf{Q} \mathbf{e} + \mathbf{e}^t \mathbf{J} \mathbf{Q} \tilde{\mathbf{R}} \mathbf{Q} \mathbf{e} - \frac{1}{4k^2} \left[(\mathbf{e}^t \mathbf{Q} \mathbf{e})^2 - (\mathbf{e}^t \mathbf{J} \mathbf{Q} \mathbf{e})^2 \right]. \quad (4b)$$

One can easily check that these equations are compatible when $m_{\text{eff}} d_{\text{eff}} \mathcal{K}^2 = \omega^2$. Incidentally, we obtain $\mathcal{K}^2 \simeq k^2 + \epsilon \mathbf{e}^t \mathbf{Q} \mathbf{e} + \epsilon^2 \mathbf{e}^t \mathbf{Q} \tilde{\mathbf{R}} \mathbf{Q} \mathbf{e} + \mathcal{O}(\epsilon^3)$, which is, as expected, the second order expansion in ϵ of the implicit wavenumber equation (B1). Note that all notations appearing in equations (3a) and (4a) are introduced in the appendix.

Results in classical multiple scattering theories are usually defined in terms of the *angular shape function* f_θ for scattering of a plane wave by a single particle. It is useful to render yet another form of the coefficients (3a) and (4a) in terms of f_θ . This is done by considering the line-like approximation: in addition to $\epsilon a^2 \ll 1$, we also require $kb \ll 1$. To render the results more tractable, the spatial distribution of particles is assumed to be isotropic and homogeneous, for which $g_{ij}(r) \equiv g(r) = H(r - b)$. This describes a non-overlapping condition; here, g denotes a *pair*-correlation function, H is the Heaviside unit function, and $b = 2a$ is the diameter of the particles. Retaining only the leading order term in kb of the multiple scattering matrix $\tilde{\mathbf{R}}$, and using the definition (C2) for f_θ , we obtain

$$\mathbf{e}^t \mathbf{Q} \tilde{\mathbf{R}} \mathbf{Q} \mathbf{e} \simeq -\frac{1}{4k^2} \mathcal{H}_0 \text{ and } \mathbf{e}^t \mathbf{J} \mathbf{Q} \tilde{\mathbf{R}} \mathbf{Q} \mathbf{e} \simeq -\frac{1}{4k^2} \mathcal{H}_\pi, \quad (5)$$

with

$$\mathcal{H}_\alpha = \frac{2}{\pi} \int_0^\pi d\theta \cot(\theta/2) \frac{d}{d\theta} \mathcal{G}_\alpha^\theta \text{ and } \mathcal{G}_\alpha^\theta = f_\theta f_{\alpha-\theta}. \quad (6)$$

By means of these approximations, we can infer the following closed-form constitutive relations

$$\frac{m_{\text{eff}}}{m} \simeq 1 + \frac{\epsilon}{2k^2} (f_0 - f_\pi) + \frac{\epsilon^2}{8k^4} \left[(\mathcal{G}_0^\pi - \mathcal{G}_0^0) - (\mathcal{H}_0 - \mathcal{H}_\pi) \right], \quad (7)$$

$$\frac{d_{\text{eff}}}{d} \simeq 1 + \frac{\epsilon}{2k^2} (f_0 + f_\pi) + \frac{\epsilon^2}{8k^4} \left[(\mathcal{G}_0^\pi - \mathcal{G}_0^0) - (\mathcal{H}_0 + \mathcal{H}_\pi) \right]. \quad (8)$$

Apart from their dependence on k and ϵ (or η), the effective dynamic parameters $(m_{\text{eff}}, d_{\text{eff}})$ given by equations (7) and (8) are all completely determined when the angular shape function f_θ for an isolated particle is known. If this scattering amplitude can be determined either analytically, numerically, or experimentally, then the effective medium equivalent to the artificial composite is *fully* described.

It is noteworthy that if one wants to study the behavior of effective parameters at high concentrations (where such expansions may not be valid) the general implicit equations detailed in appendix B should be used and/or more accurate pair correlation functions should be considered. Neither incident wave nor boundary conditions have entered yet in the above description. Consequently, the results admit several solutions corresponding to different polarization states. In the supplementary material³ (section S1), the expansions (1) and (2) (or (7) and (8)) are specialized to electromagnetic, acoustic, and elastic scattering for long wavelengths ($a \ll \Lambda$). This provides an additional check on the correctness of the results obtained in this paper. A further check on the consistency of our method is provided in the supplementary material (section S2). It is shown that the quasi-crystalline approximation is *self-consistent* and identical to coherent potential approximation [44] at least to second order in concentration, provided the effective parameters are identified as those derived in this section.

Discussion

While the limiting cases considered in the supplementary material (section S1) perform a check of the theory we have presented, they neglect some important features of the effective field method. Therefore, we address this problem numerically in order to illustrate the dynamic behavior of the effective parameters. In the following, the

³ See supplementary text for a check on the consistency of our method, and an alternative self-consistent procedure.

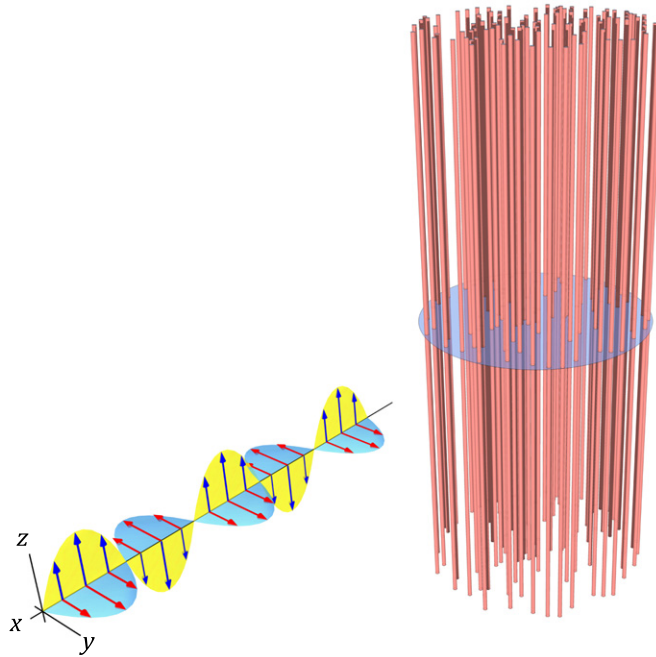


Figure 1. Sketch of the scattering of an electromagnetic wave by a circular cluster of cylindrical particles.

effective parameters (\mathbf{m}_{eff} , \mathbf{d}_{eff}) are calculated by using equations (1) and (2), together with the Percus–Yevick pair-correlation function for hard disks [45].

Example illustration

We consider a fiber bundle (or circular cluster of dielectric fibers with $\mu_0 = 1$) of effective radius r_{eff} in vacuum. A plane electromagnetic wave is incident on the fiber bundle. A sketch is shown in figure 1. There are 68 circular fibers each of radius a randomly distributed in the cluster and their volume fraction is 10.88%. The refractive index of the fibers is $n_0 (= \sqrt{\epsilon_0}) = 1.33 + 0.01i$. Exact multiple scattering simulations⁴ are compared with the effective medium model (*i.e.* equivalent homogeneous magneto-dielectric inclusion⁵ with effective parameters $(\epsilon_{\text{eff}}, \mu_{\text{eff}})$). The multiple scattering results are averaged over different realizations of the fibers locations. With 500 realizations, the maximum error between the numerical model and the effective medium results is less than 0.5%, for the two cases illustrated.

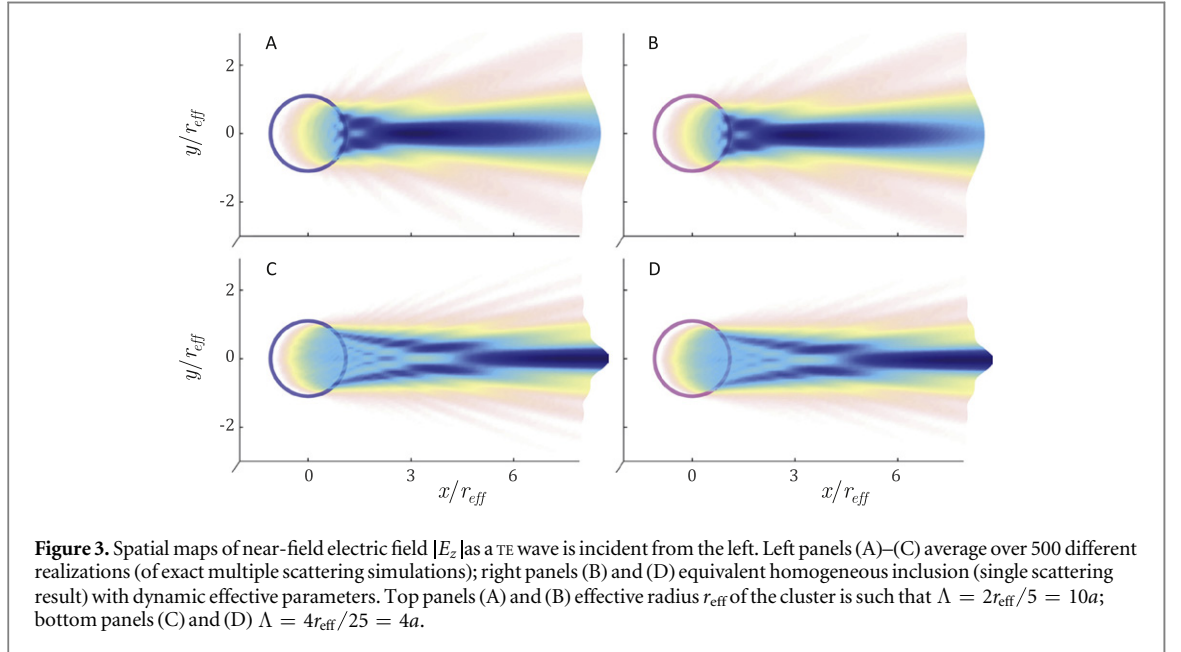
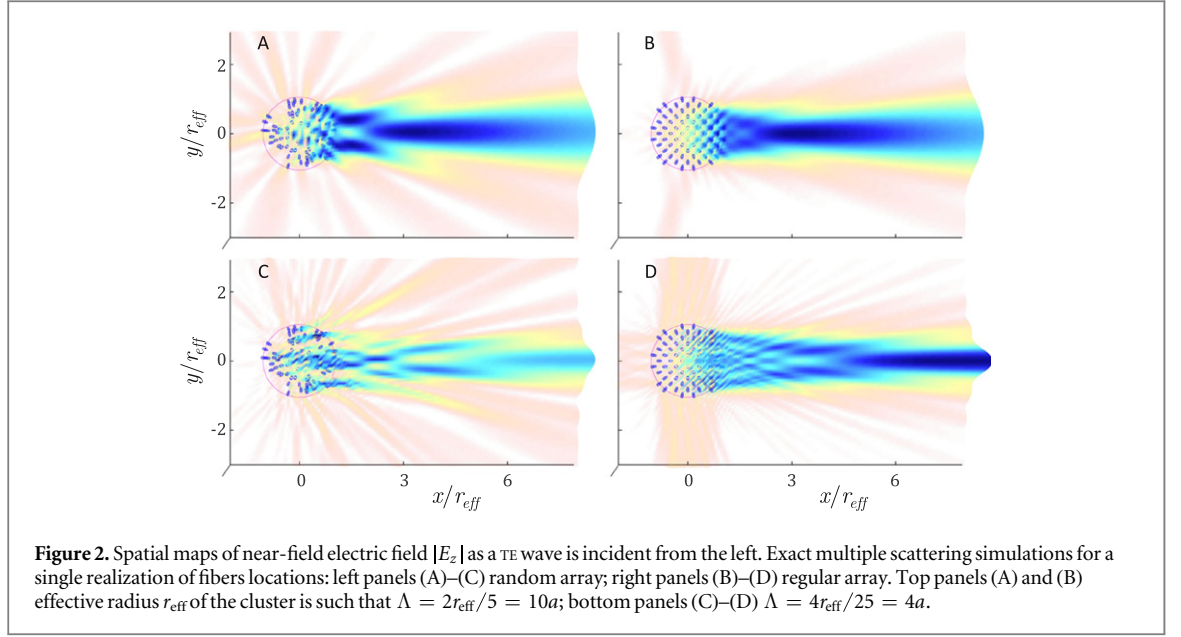
Figures 2 and 3 show the spatial maps of the near-field electric field amplitude $|E_z|$ for two different incident wavelengths, $\Lambda = 2r_{\text{eff}}/5 = 10a$ and $\Lambda = 4r_{\text{eff}}/25 = 4a$, respectively. Figure 2 illustrates the response of the coherent wave regarding the topology of the fiber-bundle. As expected, the waves are insensitive to the relative locations of the fibers for long wavelengths. This is not the same for shorter wavelengths. A comparison of the results in figure 3 indicates the agreement is excellent even for the high frequency case considered ($\Lambda = 4a$). It is particularly encouraging that the agreement is excellent even inside the circular cluster. Observe that the a regular arrangement of fibers produces a result that is closer to the effective cluster for long wavelengths, than is the result obtained with a random realization of the fibers locations. We should note that although the comparison in figure 3 is excellent, it may not always be so for other geometries of the fiber bundle. In a final section we detail various limitations and assumptions of our model and discuss other similar problems obtained previously.

Anisotropic metamaterials

It is of considerable interest to discuss the possibility of realizing anisotropic metamaterials, that is, the material parameters are not scalars but tensors, with their principle components taking different values. Different from the anisotropy property of the material itself, we shall examine anisotropy originating from geometric asymmetry and consider a random array of elliptic cylinders of material parameters $(\mathbf{m}_0, \mathbf{d}_0)$. The x - and y -axes are set in the directions of the semi-minor and semi-major axes of the elliptic cylinders, with respective radii a_x

⁴ The analytical solution to Maxwell equations for scattering by multiple parallel cylinders has been described, e.g., in [48].

⁵ Note that, as expected, our results also predict an effective magnetic permeability μ_{eff} at finite frequencies (different from that in vacuum) in a system in which both the matrix and the particles are non-magnetic.



and a_y . Due to the geometric arrangement of the elliptic cylinders and the symmetry of the scattered fields, the x - and y -directions can therefore be seen as effective principal directions. Proceeding essentially as detailed in the supplementary material (section S1), we obtain, in the quasi-static limit

$$\frac{m_{\text{eff},x}}{m} \simeq 1 + 2\phi\mathcal{M}_x + 2\phi^2\mathcal{M}_x^2, \quad (9)$$

$$\frac{m_{\text{eff},y}}{m} \simeq 1 + 2\phi\mathcal{M}_y + 2\phi^2\mathcal{M}_y^2, \quad (10)$$

$$\frac{d_{\text{eff}}}{d} \simeq 1 + \phi\mathcal{D}, \quad (11)$$

where $\phi = \pi\eta a_x a_y$ is the volume fraction of the elliptical cylinders. The coefficients \mathcal{D} and $(\mathcal{M}_x, \mathcal{M}_y)$ are given by

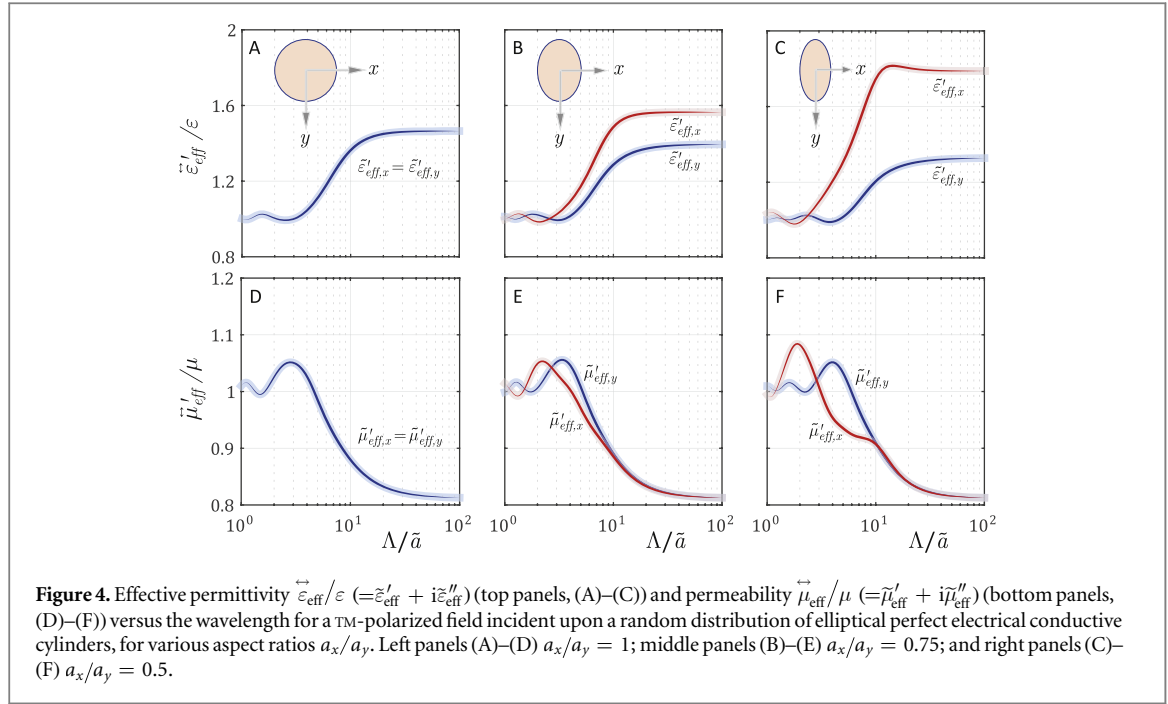


Figure 4. Effective permittivity $\tilde{\epsilon}_{\text{eff}}/\epsilon$ ($=\tilde{\epsilon}'_{\text{eff}} + i\tilde{\epsilon}''_{\text{eff}}$) (top panels, (A)–(C)) and permeability $\tilde{\mu}_{\text{eff}}/\mu$ ($=\tilde{\mu}'_{\text{eff}} + i\tilde{\mu}''_{\text{eff}}$) (bottom panels, (D)–(F)) versus the wavelength for a TM -polarized field incident upon a random distribution of elliptical perfect electrical conductive cylinders, for various aspect ratios a_x/a_y . Left panels (A)–(D) $a_x/a_y = 1$; middle panels (B)–(E) $a_x/a_y = 0.75$; and right panels (C)–(F) $a_x/a_y = 0.5$.

$$\mathcal{D} = \frac{d_0}{d} - 1 \text{ and } \begin{pmatrix} \mathcal{M}_x \\ \mathcal{M}_y \end{pmatrix} = \frac{1}{2} \frac{(m_0 - m)(a_x + a_y)}{m_0 \begin{pmatrix} a_x \\ a_y \end{pmatrix} + m \begin{pmatrix} a_y \\ a_x \end{pmatrix}}. \quad (12)$$

Observe that if $a_x = a_y$ ($=a$, i.e., circular cross section) then $\mathcal{M}_x = \mathcal{M}_y$ ($=\mathcal{M}$, see supplementary material, section S1), and $m_{\text{eff},x} = m_{\text{eff},y}$. The results of equations (9)–(11) show that only the effective property $\tilde{\mathbf{m}}_{\text{eff}}$ is a tensor with principal components $m_{\text{eff},x}$ and $m_{\text{eff},y}$, whereas \mathbf{d}_{eff} is a scalar. This is consistent with results obtained recently in [28], for electromagnetic waves in the quasi-static limit. However, these results should be consumed with prudence. We show that, in general, both $\tilde{\mathbf{m}}_{\text{eff}}$ and \mathbf{d}_{eff} are tensors for arbitrary frequency and wavelength. To see this more clearly, let us consider the scattering of a TM wave by perfect electric conductive elliptic cylinders in vacuum⁶. From table 1, we infer that (\mathbf{m}, \mathbf{d}) corresponds to (ϵ, μ) , for TM waves; appropriate identifications the resulting effective medium are implied. Figure 4 shows the effective permittivity $\tilde{\epsilon}_{\text{eff}}$ and permeability $\tilde{\mu}_{\text{eff}}$ tensors. Only the real part of these parameters is presented for brevity. The volume fraction ϕ is fixed and equal to $6\pi\%$. It should be noted that the actual concentration $\phi = \pi\eta a_x a_y$ cannot exceed a_x/a_y in order to be consistent with our model, so that $\phi = \pi\eta a^2 \leq 1$ when $a_x = a_y$ ($=a$). The figure is intended to illustrate the variations of $\tilde{\epsilon}_{\text{eff}}/\epsilon$ and $\tilde{\mu}_{\text{eff}}/\mu$ as the wavelength Λ/\tilde{a} varies on the horizontal axis, for several aspect ratios a_x/a_y ; here, $\tilde{a} = \sqrt{a_x a_y}$ is the geometric mean of the semi-minor and semi-major axes, a_x and a_y . Observe that in the quasi-static limit, for $\Lambda > 10\tilde{a}$, where currently available model will be adequate, the principal components of $\tilde{\mu}_{\text{eff}}$ are visibly equal, i.e., $\mu_{\text{eff},x} \simeq \mu_{\text{eff},y}$, regardless of the ratio a_x/a_y . This is as expected, given equation (11). It is interesting that for shorter wavelengths ($\Lambda < 10\tilde{a}$), $\mu_{\text{eff},x}$ and $\mu_{\text{eff},y}$ become increasingly distinct as the ratio a_x/a_y decreases from 1 to 0.5, an effect not predicted by the existing literature. This suggests a new route to the design of metamaterials with controllable anisotropic effective properties.

Conditions of applicability

The results in figures 2 and 3 support the reliability of the effective material parameters resulting from the *supra*-classical dynamic homogenization procedure reported here. Note however that, although not apparent in the results, there is an approximation involved in replacing a finite-size heterogeneous composite with its homogenized equivalent, in addition to the reliability of the homogenization procedure (which ignores transition region complications at the interface $|x| \leq a$ [41]). In practical terms, it means that for a finite sample of the random composite the applicability of dynamic homogenization not only depends upon the frequency under consideration but also upon the phase of the composite at the boundary of the sample. An effort to quantify such an approximation is described in [46]. Here, the approximation results from truncating interfaces

⁶ This is equivalent to solving the Neumann boundary condition. In anti-plane elasticity, this condition corresponds to a cylindrical cavity with stress-free surface.

of a finite (or semi-infinite) 1-D periodic composite, the later being replaced with what are essentially its effective dynamic properties in the infinite Bloch-wave domain. Other questions will need to be answered relating to the shape and size of the scattering boundary, the effect of increasing the number of particles, and how many realizations are required to determine both the near- and far-fields accurately. It is expected that as the bounded area increases, so does the uncertainty of the calculated field. An investigation in this direction is beyond the scope of the paper, however we refer the reader to the comprehensive numerical analysis (based on the quasi-crystalline approximation) reported in [47, 52]. The later references should come with a warning, as their analysis contains the implicit (and incorrect [53]) assumption that the dielectric permittivity is the only quantity of interest.

Finally, let us note that the effective material parameters derived in here (which are tensor values for anisotropic media) are not necessarily tied to the physical material parameters of any of the individual elements of the metamaterial. A rather critical survey discussing the link and the difference between these two concepts (i.e. effective versus characteristic material parameters), particularly for the case of Maxwell's equations, is presented in [54]. As evidenced in this survey and references therein, homogenization theories continue to attract attention and even controversy. It appears, from considering exact reflection coefficients at oblique incidence (if one assumes that Fresnel-like formulae are always valid), that any effective material parameters that can be introduced in any theory would depend on the angle of incidence; broadly speaking, they would depend on the type of illumination. This means that these effective properties do not necessarily relate solely to the bulk properties of the material itself; they can involve the material and the type of illumination. Relevant considerations in this direction are presented for periodic composites in [55, 56]. A retrieval method extended to the arbitrary orientations of the principal axes of anisotropy and oblique incidence was presented in [57]. A discussion regarding modelling of the coherent wave propagation from the knowledge of the material properties along the principal axes only is elaborated in [58].

To summarize, a self-consistent multiple scattering approach, which enables the dynamic homogenization of metamaterials in two-dimensions, is developed. The quasi-crystalline approximation is employed to break the hierarchy of increasing conditional probability densities, but otherwise the treatment is exact. In particular, the effective wavenumber and the effective impedance is obtained. These characteristics can then be used to determine the effective constitutive parameters of the homogenised material. Whether the resulting effective parameters represent a true bulk property of the metamaterial in the dynamic range is yet to be determined. The two natural approximations—dilute media and low frequency approximations—show consistency, and, moreover, the quasi-static limit gives results reminiscent of the laws of Maxwell Garnett [49], Ament [50], and Kuster and Toksöz [51], respectively for electromagnetic, acoustic, and elastic material parameters (see supplementary material, section S1, for more details). The entire analysis described in this work is germane for alternative analytical procedures based on other scattering operators \mathbf{Q} for an isolated particle. As shown in the supplementary material (section S2), a fully self-consistent procedure may be based on a new kind of isolated scatterer problem. We have shown that the coherent potential approximation, used in many previous works, is only an approximation of this procedure to the first order in the concentration of particles.

The theory provided here offers exciting opportunities for researchers in different communities, ranging from seismic waves to the entire field of ultrasound research, and spanning radio frequency and optical engineering. In particular, metamaterial modelling in optics, physical acoustics, and condensed matter physics may benefit from a rigorous, compact model for estimating more accurate and anisotropic effective medium parameters that homogenize artificial media.

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Appendix A. Governing equation

There are many examples of wave equations in the physical sciences, characterized by oscillating solutions that propagate through space and time while, in lossless media, conserving energy. Examples include the scalar wave equation (e.g., pressure waves in a gas), Maxwell's equations (electromagnetism), Schrödinger's equation (quantum mechanics), elastic vibrations, and so on. From a mathematical viewpoint, all of these share certain common features. In the following, we shall briefly identify the similarities between three types of such waves, in two-dimensions: electromagnetic waves, anti-plane elastic waves, and acoustic waves. Electromagnetic waves are quite different from acoustic and elastic waves in that they can travel through vacuum. However, from an

algebraic perspective, all three types of waves can be described by a unique scalar equation and hence these disparate phenomena can be studied simultaneously. The prototype problem consists of the wave equation at fixed angular frequency ω

$$\nabla \cdot (\mathbf{m}^{-1}(\mathbf{r}) \nabla \psi(\mathbf{r})) + \omega^2 \mathbf{d}(\mathbf{r}) \psi(\mathbf{r}) = 0, \quad (\text{A1})$$

where $c = 1/\sqrt{\mathbf{m}\mathbf{d}}$ is the phase velocity of the wave for some parameters $\mathbf{m}(\mathbf{r})$ and $\mathbf{d}(\mathbf{r})$ of the medium. In the above, the scalar wavefunction $\psi(\mathbf{r})e^{-i\omega t}$ corresponds to some physical field.

The problem considered here is reduced to points in the x - y plane (i.e. the cross section plane of our scattering geometry), which in polar coordinates are $\mathbf{r} = (r, \theta)$; here, θ is measured from the positive x -axis. Let us first consider the two important modes for electromagnetic wave propagation: the transverse electric field and the transverse magnetic field. These modes are closely analogous to anti-plane shear in elastodynamics and to acoustic waves. Let us assume the medium is isotropic and has dielectric permittivity ε and magnetic permeability μ that are independent of z . A transverse magnetic (TM) field is a special solution of the Maxwell's equations that has the form $\mathbf{H} = \psi(x, y)\mathbf{i}_z$, and the electrical field $i\omega\varepsilon\mathbf{E} = -\nabla \times \mathbf{H} = \frac{\partial\psi}{\partial y}\mathbf{i}_x - \frac{\partial\psi}{\partial x}\mathbf{i}_y$. A transverse electric (TE) field is another special solution of the Maxwell's equations that has the form $\mathbf{E} = \psi(x, y)\mathbf{i}_z$, whereby the magnetic field is given by $i\omega\mu\mathbf{H} = \nabla \times \mathbf{E} = \frac{\partial\psi}{\partial x}\mathbf{i}_y - \frac{\partial\psi}{\partial y}\mathbf{i}_x$. Observe that simple knowledge of the scalar wavefunction ψ suffices for the determination of the x and y components of the electric and magnetic fields, for the two polarizations. Hence, equation (A1) is the governing wave equation for electromagnetic waves provided that $(\mathbf{m}, \mathbf{d}, \psi) \leftrightarrow (\varepsilon, \mu, H_z)$ for TM waves, and $(\mathbf{m}, \mathbf{d}, \psi) \leftrightarrow (\mu, \varepsilon, E_z)$ for TE waves. The general solution independent of z is a superposition of the TE and TM solutions. This can be seen by observing that the Maxwell's equations decouple under this condition and a general solution can be written as $(H_x, H_y, H_z) = (H_x, H_y, 0) + (0, 0, H_z)$, where the second term represents the TM solution. The first term is of course the TE solution because $\nabla \times (H_x, H_y, 0) = (0, 0, \frac{\partial}{\partial y}H_x - \frac{\partial}{\partial x}H_y)$, which implies $E_x = E_y = 0$ as expected. Let us now consider the case of anti-plane shear strain which is a special state of deformation where the displacement field is given by $\mathbf{u} = \psi(x, y)\mathbf{i}_z$. This is an out-of-plane mode of deformation and is analogous to transverse electromagnetic wave propagation. In the linear regime, an isotropic elastic medium is characterized by its density ρ and the Lamé elastic constants $G \equiv \mu$ and λ : G is the shear modulus (notation used to distinguish from the permeability μ employed in electromagnetics) and $\lambda = \kappa - G$ where κ is the two-dimensional bulk modulus. Hence, equation (A1) is the wave equation for anti-plane shear provided that $(\mathbf{m}, \mathbf{d}, \psi) \leftrightarrow (1/G, \rho, u_z)$. Shear waves that satisfy this equation are also called s_H (shear horizontal) waves, particularly in seismology. Finally, let us consider the acoustic wave propagation in an isotropic medium. For an inviscid fluid or gas, the shear modulus G is zero—and λ is just the bulk modulus. In this case, replacing ψ with the pressure $p = -\kappa \nabla \cdot \mathbf{u}$, we obtain precisely the acoustic wave equation (A1) for P (or compressional) waves if $(\mathbf{m}, \mathbf{d}) \leftrightarrow (\rho, 1/\kappa)$.

In essence, the solutions of the three problems considered will lead to similar conclusions if we make the appropriate interpretation of the quantities involved.

Appendix B. Effective field method

Suppose that discrete particles of cylindrical geometry are randomly and uniformly distributed in a half-space defined by $\{x > 0\}$. The particles need not be circular, provided that each of them can be contained in a circumscribing circular surface of radius a_j (with an axis of revolution parallel to the z -axis); their number density is η_j . Both the particles and the matrix are made of isotropic materials. Let a plane wave $\psi = \exp[i(kx - \omega t)]$ of unit amplitude propagate with wavenumber k in the matrix along the x -direction. When this wave propagates in the composite material, multiple scattering occurs. Either propagation or diffusion, or a combination of the two phenomena is observed, depending on the frequency as well as on the geometrical and material properties of the composite. Assuming that propagation occurs, one can describe the coherent wave motion in the composite by a complex-valued wavenumber \mathcal{K} . The fundamental equation for configurational averages of the exciting and total fields for scalar wavefunctions has been derived in detail in [40, 41, 52, 59]. The quasi-crystalline approximation [60] is used to truncate the hierarchy of equations (Foldy-Lax hierarchy) so that only the correlation between every two particles is considered. We obtain the implicit dispersion equation for the effective wavenumber \mathcal{K} of the coherent wave $\exp[i(\mathcal{K}x - \omega t)]$

$$\mathcal{K}^2 = k^2 + \sum_j \epsilon_j \mathcal{F}(a_j), \quad (\text{B1})$$

where $\epsilon_j = 4\pi\eta_j$, and the effective scattering amplitude \mathcal{F} is given by

$$\mathcal{F}(a_j) = \mathbf{e}^t (\mathbf{Q}_j^{-1} - \sum_i \epsilon_i \mathbf{R}_{ij})^{-1} \mathbf{e}, \quad (\text{B2})$$

$\mathbf{e} = (1, 1, \dots)^t$ is a constant unit vector. The shorthand notation $\mathbf{R}_{ij} \equiv \mathbf{R}(b_{ij})$ and $\mathbf{Q}_j \equiv \mathbf{Q}(a_j)$ has been used. The infinite square matrices $\mathbf{R}(b_{ij})$ and $\mathbf{Q}(a_j)$, have elements

$$\mathcal{R}_{m\nu}(b_{ij}) = \frac{\mathcal{P}_{n-\nu}(\mathcal{K}b_{ij}) - 1}{\mathcal{K}^2 - k^2} + \mathcal{N}_{n-\nu}(\mathcal{K}b_{ij}), \quad (\text{B3})$$

where $b_{ij} \geq a_i + a_j$, and

$$Q_{m\nu}(a_j) = \frac{1}{i\pi} \delta_{m\nu} T_n(a_j). \quad (\text{B4})$$

Here, δ_{ij} denotes the Kronecker delta, and \mathcal{P}_ℓ and \mathcal{N}_ℓ are given by

$$\begin{aligned} \mathcal{P}_\ell(z) &= \frac{i\pi}{2} \left[z H_\ell^{(1)}(x) \frac{d}{dz} J_\ell(z) - x J_\ell(z) \frac{d}{dx} H_\ell^{(1)}(x) \right], \\ \mathcal{N}_\ell(z) &= \frac{i\pi}{2} \int_{b_{ij}}^{\infty} dr \left[g_{ij}(r) - 1 \right] r H_\ell^{(1)}(kr) J_\ell(zr/b_{ij}), \end{aligned}$$

where J_ℓ and $H_\ell^{(1)}$ are the cylindrical Bessel and Hankel functions, respectively, and $x = kb_{ij}$. The function g_{ij} is the cross-pair distribution function of two particle species (with sizes a_i and a_j), and satisfies the non-overlapping condition: $g_{ij}(r) = 0$ for $r < b_{ij}$; also, if the distance between particles tends to infinity, then the correlation between their locations disappears, i.e., $\lim_{r \rightarrow \infty} g_{ij}(r) = 1$.

The scattering coefficients $T_n(a_j)$ in equation (B4) depend on frequency, size a_j , as well as on the properties of the particle and those of the matrix material; they are evaluated by imposing appropriate boundary conditions at $r = a_j$.

Equation (B1) follows directly from a Lorentz–Lorenz-type law, and is an exact expression for the effective wavenumber, subject to the quasi-crystalline approximation. It is of interest to note how various physical aspects are embedded in this equation. The scattering matrix \mathbf{Q} describes the response of a single particle to a plane incident harmonic wave with wavenumber k , and contains all the scattering behavior in terms of particle geometry and physical parameters. The effective wavenumber \mathcal{K} only appears in the matrix \mathbf{R} , which is defined by the spatial arrangements of particles, and accounts for multiple scattering. Should the distribution of particles be regular, the quasi-crystalline approximation is exact, in which case the multiple-scattering matrix \mathbf{R} can be reduced to a well known lattice sum.

The theory described above is now complete insofar as behavior within the medium is concerned. It is also of interest, however, to calculate the effective impedance \mathcal{Z} , which defines the reflectivity of the half-space $\{x > 0\}$ —a quantity which may be measured directly. Following the derivations in [37, 41], the coherent reflected field $\langle \psi \rangle = \Re \exp(-ikx)$ at the half-space boundary can be obtained explicitly, with the reflection coefficient defined as

$$\Re = - \frac{\sum_j \epsilon_j \mathcal{F}_\pi(a_j)}{4k^2 + \sum_j \epsilon_j \mathcal{F}_0(a_j)}. \quad (\text{B5})$$

Here, \Re represents the average (coherent) back-scattered amplitude at normal incidence in the domain $\{x < 0\}$. The effective scattering amplitudes, \mathcal{F}_0 and \mathcal{F}_π , correspond to coherent waves scattered in the forward and backward directions, respectively, and are given by

$$\mathcal{F}_0(a_j) = \mathbf{e}^t \mathbf{Q}_j \mathbf{v}_j \text{ and } \mathcal{F}_\pi(a_j) = \mathbf{e}^t \mathbf{J} \mathbf{Q}_j \mathbf{v}_j, \quad (\text{B6})$$

where $\mathbf{J} = \{\delta_{m\nu} \cos n\pi\}$ is a diagonal infinite matrix. The infinite eigenvector \mathbf{v}_j , associated with the wavenumber equation, follows from an Ewald–Oseen-type extinction theorem, with the result

$$\mathbf{v}_j = \frac{2k}{\mathcal{K} + k} \left(\mathbf{I} - \sum_i \epsilon_i \mathbf{Q}_j \mathbf{R}_{ij} \right)^{-1} \mathbf{e}, \quad (\text{B7})$$

where \mathbf{I} is a unit infinite matrix. Martin [61] has obtained a formula for \Re for obliquely incident waves on a half-space of circular scatterers; it can be shown that at normal incidence the result in his equation (39) gives agreement with equation (B5). The behavior of the fields across interfaces was also examined in [61–63]; it was found that the fields themselves are continuous but the slopes are discontinuous. Using the estimate for the slope discontinuity, effective constitutive parameters can be derived, as shown in [61, 63]. Equation (B5) is an exact

formula for the reflection coefficient⁷; it can be used to determine effective parameters (\mathbf{m}_{eff} , \mathbf{d}_{eff}) uniquely. It is often assumed that the effective medium corresponding to the distribution of particles may be described as a homogeneous medium from the standpoint of coherent wave propagation—the homogenized equivalent having the effective dynamic properties of the composite. In the following, we shall use this analogy, whereby the reflection coefficient \mathfrak{R} at the interface between the homogeneous medium and the homogenized equivalent, may be written (as is standard) in terms of impedances (resulting in a Fresnel-like formula)⁸. Then, equating the result with equation (B5), the effective impedance \mathcal{Z} can be explicitly calculated. As expected, the effective impedance is different for different polarizations. Two cases are possible, with the following results:

$$\mathcal{Z}^{\text{m}} = \mathcal{Z}/z \text{ and } \mathcal{Z}^{\text{d}} = z/\mathcal{Z}, \quad (\text{B8})$$

where

$$\mathcal{Z} = z \frac{4k^2 + \sum_j \epsilon_j \left[\mathcal{F}_0(a_j) - \mathcal{F}_\pi(a_j) \right]}{4k^2 + \sum_j \epsilon_j \left[\mathcal{F}_0(a_j) + \mathcal{F}_\pi(a_j) \right]}. \quad (\text{B9})$$

Here, $z = \sqrt{\mathbf{m}/\mathbf{d}}$ is the impedance of the matrix; the superscripts ‘m’ and ‘d’ correspond to different physical situations, as we shall see below. We can now state our most general expressions for the effective dynamic constitutive parameters (\mathbf{m}_{eff} , \mathbf{d}_{eff}),

$$\frac{\mathbf{m}_{\text{eff}}}{\mathbf{m}} = \frac{\mathcal{K}}{k} \mathcal{Z}^{\text{m}} \text{ and } \frac{\mathbf{d}_{\text{eff}}}{\mathbf{d}} = \frac{\mathcal{K}}{k} \mathcal{Z}^{\text{d}} \quad (\text{B10})$$

where (\mathcal{Z}^{m} , \mathcal{Z}^{d}) are defined in equations (B8)–(B9). Observe that, by using the definition,

$$\mathcal{K} = k + \frac{1}{2k} \sum_j \epsilon_j \mathcal{F}_0(a_j) \quad (\text{B11})$$

in equation (B10), the resulting parameters (\mathbf{m}_{eff} , \mathbf{d}_{eff}) can be expressed explicitly in terms of the effective forward and back-scattering shape functions, \mathcal{F}_0 and \mathcal{F}_π .

To conclude this section, we consider the line-like approximation of the constitutive parameters (B10). For this, the size of the particles is assumed small compared to the incident wavelength ($a_j \ll \Lambda$). At leading order, the single-scattering operator \mathbf{Q} is compact and has only three eigenvalues of finite size (related to terms with $n = 0, \pm 1$). Furthermore, the infinite multiple-scattering operator \mathbf{R} is reduced to a rank 3 matrix. Omitting the details, we find for circular cylinders (with $T_1 = T_{-1}$),

$$\frac{\mathbf{m}_{\text{eff}}}{\mathbf{m}} \simeq \frac{k^2 + \frac{1}{i\pi} \sum_j \epsilon_j T_1(a_j)}{k^2 - \frac{1}{i\pi} \sum_j \epsilon_j T_1(a_j)}, \quad (\text{B12})$$

$$\frac{\mathbf{d}_{\text{eff}}}{\mathbf{d}} \simeq 1 + \frac{1}{i\pi k^2} \sum_j \epsilon_j T_0(a_j). \quad (\text{B13})$$

It can be shown that in the quasi-static limit ($a_j \ll \Lambda_0$) the effective property \mathbf{m}_{eff} is reminiscent of the laws of Maxwell Garnett [64], Ament [50], and Kuster and Toksöz [51], in two-dimensions, respectively for electromagnetic, acoustic, and elastic material parameters. (This is further described in the supplementary material, section S1.) On the other hand, the effective property \mathbf{d}_{eff} reduces to the simple and inverse rules of mixtures, depending on the physical model under consideration, and as seen from equation (B13) is linear in ϵ_j .

Appendix C. Explicit second order approximations

At low concentrations ($\epsilon_j a_j^2 \ll 1$), the dispersion equation is explicit, and reduces to the well-known formula [40]

$$\mathcal{K}^2 \simeq k^2 + \sum_j \epsilon_j f_0(a_j), \quad (\text{C1})$$

where the forward-scattering amplitude f_0 is given by $f_0(a_j) = \mathbf{e}^t \mathbf{Q}_j \mathbf{e}$. More generally, the angular shape function f_θ for each particle is defined, in terms of Fourier series, as⁹

⁷ Note however that during the derivation, complications in the transition region $-a \leq x \leq a$ near and on both sides of the interface have been ignored [41].

⁸ For instance, for acoustic waves, $\mathfrak{R} = (\mathcal{Z} - z)/(\mathcal{Z} + z)$; this result implies the continuity of pressure and normal velocity at the interface; for anti-plane elastic waves, $\mathfrak{R} = -(\mathcal{Z} - z)/(\mathcal{Z} + z)$; here, the continuity of the out-of-plane displacement and the corresponding stress are implicit. Similar results in electromagnetics are known as Fresnel relations (for TE and TM waves).

⁹ Note the shorthand notation $\sum_n = \sum_{n=-\infty}^{\infty}$ is used throughout.

$$f_\theta(a_j) = \frac{1}{i\pi} \sum_n T_n(a_j) e^{in\theta}. \quad (C2)$$

An expansion of the dispersion equation (B1) to the second order in concentration results in

$$K^2 \simeq k^2 + \sum_j \epsilon_j \mathbf{e}^t \mathbf{Q}_j \mathbf{e} + \sum_{i,j} \epsilon_i \epsilon_j \mathbf{e}^t \mathbf{Q}_i \tilde{\mathbf{R}}_{ij} \mathbf{Q}_j \mathbf{e} + \mathcal{O}(\epsilon_i \epsilon_j \epsilon_k), \quad (C3)$$

where the matrix $\tilde{\mathbf{R}}_{ij} \equiv \tilde{\mathbf{R}}(b_{ij}) = \lim_{K \rightarrow k} \mathbf{R}_{ij}$ and has elements

$$\tilde{\mathcal{R}}_{\nu\nu}(b_{ij}) = \mathcal{N}_\ell(x) + \frac{i\pi}{4k^2} \left[(\ell^2 - x^2) J_\ell(x) H_\ell^{(1)}(x) - x^2 \frac{d}{dx} J_\ell(x) \frac{d}{dx} H_\ell^{(1)}(x) \right], \quad (C4)$$

with $\ell = n - \nu$, and $x = kb_{ij}$. Note that for spatially uncorrelated particles, $\mathcal{N}_\ell(x) = 0$.

For the effective impedance of equation (B9), at first order in concentration, we have

$$\mathcal{Z} = z - \frac{1}{2k^2} \sum_j \epsilon_j f_\pi(a_j), \quad (C5)$$

where the back-scattering amplitude f_π is given by $f_\pi(a_j) = \mathbf{e}^t \mathbf{J} \mathbf{Q}_j \mathbf{e}$. The second order approximation is too long to warrant including here. For completeness, we also give the following results, in terms of Fourier series,

$$\mathbf{e}^t \mathbf{Q}_i \tilde{\mathbf{R}}_{ij} \mathbf{Q}_j \mathbf{e} = \frac{i}{\pi^2} \sum_{n,v} \tilde{\mathcal{R}}_{\nu\nu}(b_{ij}) T_n(a_i) T_\nu(a_j), \quad (C6)$$

$$\mathbf{e}^t \mathbf{J} \mathbf{Q}_i \tilde{\mathbf{R}}_{ij} \mathbf{Q}_j \mathbf{e} = \frac{i}{\pi^2} \sum_{n,v} (-1)^n \tilde{\mathcal{R}}_{\nu\nu}(b_{ij}) T_n(a_i) T_\nu(a_j). \quad (C7)$$

These expressions can be easily approximated in the low frequency limit by observing that, to leading order in (kb_{ij}) , and for uncorrelated particles, $\tilde{\mathcal{R}}_{\nu\nu} \cong |n - \nu|/2k^2$. The results obtained here have been used to derive the analytic formulae presented in the main text.

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